MAGNETIC AND TRANSPORT PROPERTIES OF $^{154}Sm_{0.5}Sr_{0.5}MnO_3$ MANGANITE

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We report here the investigations of transport and magnetic properties of the $x = 0.5$ manganite which include the resistivity, magnetization as well as nonlinear response to ac-field measurements.

$^{154}Sm_{0.6}Sr_{0.4}MnO_3$ investigated in Ref. [1] was used to prepare $^{154}Sm_{0.5}Sr_{0.5}MnO_3$. Mixed powder of the initial sample added with adequate amounts of $SrCO_3$ and MnO_2 was treated at 1270 K for 48 hours with a few intermediate grinding to complete decarbonation. The obtained powder was pressed into the form of pellets and a synthesis was carried out at 1470 K and then at 1770 K for 12 hours. After it, the sample was slowly cooled down to 1070 K with a rate of 5 K/min followed by a quenching performed down to room temperature. A cation composition was determined by an energy dispersive spectroscopy (EDS) analysis on an analyser mounted on the base of the three electron microscopes. A highly homogeneous distribution of cations was found. The chemical analysis and X-ray diffraction showed also a stoichiometric compound.

Resistance and magnetoresistance in $H = 7$ T magnetic field were measured by a standard four-probe technique at cooling from 300 K to 5 K using $2\times2\times10$ mm samples. The magnetization M was registered with a vibrating sample magnetometer under $H = 1.45$ T in heating after zero-field cooling from 300 K down to 4.2 K. The measurements of the second harmonic of the longitudinal component of magnetization M_2 were performed in parallel dc- and ac-harmonic magnetic fields $H + h \sin\omega t$ ($h \leq 35$ Oe, $f = \omega/2\pi = 15.7$ MHz). An installation and a method of separation of the M_2-phase components have been described previously [2].

Resistivity $\rho(T)$ exhibits a maximum at 50 K, and reveals a metallic behavior below it (see Fig.1a). As the insert in Fig.1a shows, polaronic character of the $\rho(T)$ occurs down to 150 K. In a range 150 K ÷ 50 K, $\rho(T)$ increases with decreasing temperature, but lies below the polaronic conductivity. It is the range where according to neutron data the structural phase separation develops, starting from $T \approx 150$ K with an approximate temperature of transition $T_{st} \approx 135$ K [3]. An insulator to metal transition coincides approximately with completion of this phase separation. Besides, an analysis of the magnetic contributions to neutron diffraction has shown that the ground state is a mixture of the two magnetic phases: ferromagnetic (F) (below T_{st}) and A-type antiferromagnetic (AF-A) at $T_N \approx T_{st} \approx 135$ K. The F moment $M_F = 1.9(1)$ μ_B/Mn at T = 1.5 K and volume fraction ($V_{FI} \approx 63\%$) of F phase (for the most probable scenario: one magnetic phase in each structural phase) were also found in Ref.3. There is a reasonable correspondence between the volume fraction (V_{FI}), magnetization of the F phase and the bulk magnetization value obtained at 4.2K ($1.5\mu_B / V_{FI} \approx 2.38$ μ_B/Mn, see insert in Fig.1b). Note that the origination of the F state by a jump that starts at $T_C \approx 130$ K is found in the M_2 measurements discussed below. The metallic behavior of the sample can be attributed to the F phase since the AF-A phase is expected to be insulating. This is supported by the single crystal properties of this compound which suggest the AF insulating ground state [4,5]. In addition, a polycrystalline $Sm_{0.45}Sr_{0.55}MnO_3$ also exhibits the homogeneous insulating ground state with the crystal and magnetic structures coinciding with those of phase II [6]. The small value of the F moment ($M_F \approx 1.9$ μ_B/Mn at T = 1.5 K) suggests the presence of a magnetic disordering in the ground state. This phenomenon may be attributed to a spin-glass state that coexists with the F states. The spin-glass phase may be an intermediate one between the AF-A and F structures due to a competition of the F and AF couplings of the F-ordered

ab-planes. A similar weakly ferromagnetic spin-glass state was observed in $Pr_{1-x}Ca_xMnO_3$ at $x = 0.3$ where this state coexisted with an AF-pseudo-CE phase [7,8].As it has been explained above, the metallic behavior is related to the F phase. The metallization occurs below $T_{IM} \approx 50K$ when this phase forms a well conductive percolative net. The T_{IM} is considerably less than $T_C \approx 130$ K that can be related to the following peculiarities of the transition in our compound. First, the F ordering is accompanied by reducing fraction of the F phase due to the structural transition. Second, the originating F phase exhibits the coherent JT distortions that promote a localization of the carriers. At last, the new insulating phase II develops in the powder sample at the structural transition. It is quite possible, therefore, that this phase begins to form in the vicinity of the grain boundaries, preventing metallization of the intergrain contacts. These negative factors have to be overcame by increasing magnetization of the F phase that occurs at the relative low T_{IM} when the magnetization becomes high enough.
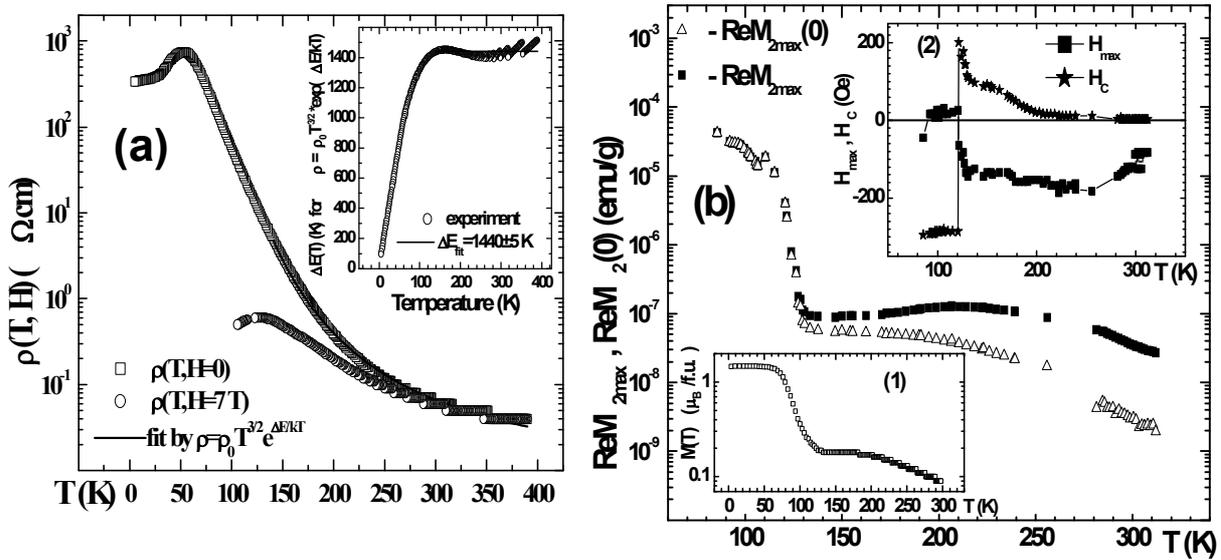

Fig.1. Temperature dependencies of resistivity and parameters of the second harmonic of magnetization. Panel (a) shows ρ(T) at H = 0 (open squares), H = 7 T (open circles) and fit the former by the polaronic law (solid line). Insert displays the resistivity at H = 0 and its fit in other coordinates. Panel (b) presents values ReM₂(H) in maximum (full squares) and at H = 0 (open triangles) vs. T. Insert (1) displays T- dependence of the magnetization. Insert (2) shows the H-position of ReM₂(H) maximum (full squares) and coercive field H_C (full stars) vs. T.

The previous measurements of $\rho(T)$ in the powder sample of the same composition show also the metallic behavior at low temperatures [9]. However, this sample reveals an essentially larger conductivity at all the temperatures. Since these samples were prepared by the different methods these observations demostrate a tendency to the low temperature metallization for the half-doped powder Sm-Sr manganites. Wide variations in the resistivity of samples synthesized by the different methods are likely due to the different synthesis procedures used for preparing these samples as a detailed study of a similar polycrystal $La_{0.5}Ca_{0.5}MnO_3$ shows [10,11]. In this manganite with metal ground state, a grain size controlling a fraction of the F phase in the ground state (9% ÷ 84%) depends substantially on the thermal treatments. It leads, first of all, to the strongly sample dependent transport properties with a tendency to an increasing of the resistivity with increasing a grain size. This can be the reason for the difference in the transport behavior of the powder and single crystalline forms of our manganite. The latter was found to be an insulator and revealed the $M(T)$ dependence suggesting a pure AF spin structure in the magnetically ordered state [4,5].

The resistivity has the rather large maximal value that is reduced considerably by the magnetic field exhibiting the strong colossal magnetoresistance effect frequently observed in phase separated manganites. In these materials a magnetic field enforces not only the alignment of the magnetic moment of the F phase. When a magnetic field of 7 T is applied during cooling, it can effect the balance between the coexisting phases by increasing strongly the amount of the F phase [10-12] that gives the well conductive state.

The peculiarities of the magnetic transition in the system with $T_C \sim T_N \approx T_{st} \approx 135$ K can also account for some difference between the magnetization measurements, neutron diffraction and nonlinear response data. The $M(T)$ dependence registered on heating at $H = 1.45$ T after zero-field cooling is similar to that of a ferromagnetic with a continuous transition at $T^*_C \sim 100$ K (insert1 in Fig.1b). As it is explained above, the neutron diffraction $M_F(T)$ data obtained also on heating, but at zero field, allows one to establish the F ordering at 110K. This result correspond to the ReM_2 measurements that reveal a jump in the spontaneous magnetization developing at 130 K (see Fig.1b). Such a behavior suggests, at least, a weak first order transition at $T_C \approx 130$ K. This transition (H is near to zero) does not generally speaking occur in the relatively large $H=1.45$T. In addition, the volume of the F phase begins to decrease at cooling that reduces a low border of a sharp changing in the $M(T)$ dependence. These factors are likely to account for the similarity of the $M(T)$ dependence with that of continuous transition characterized by effective $T_C^* \sim 100$ K.
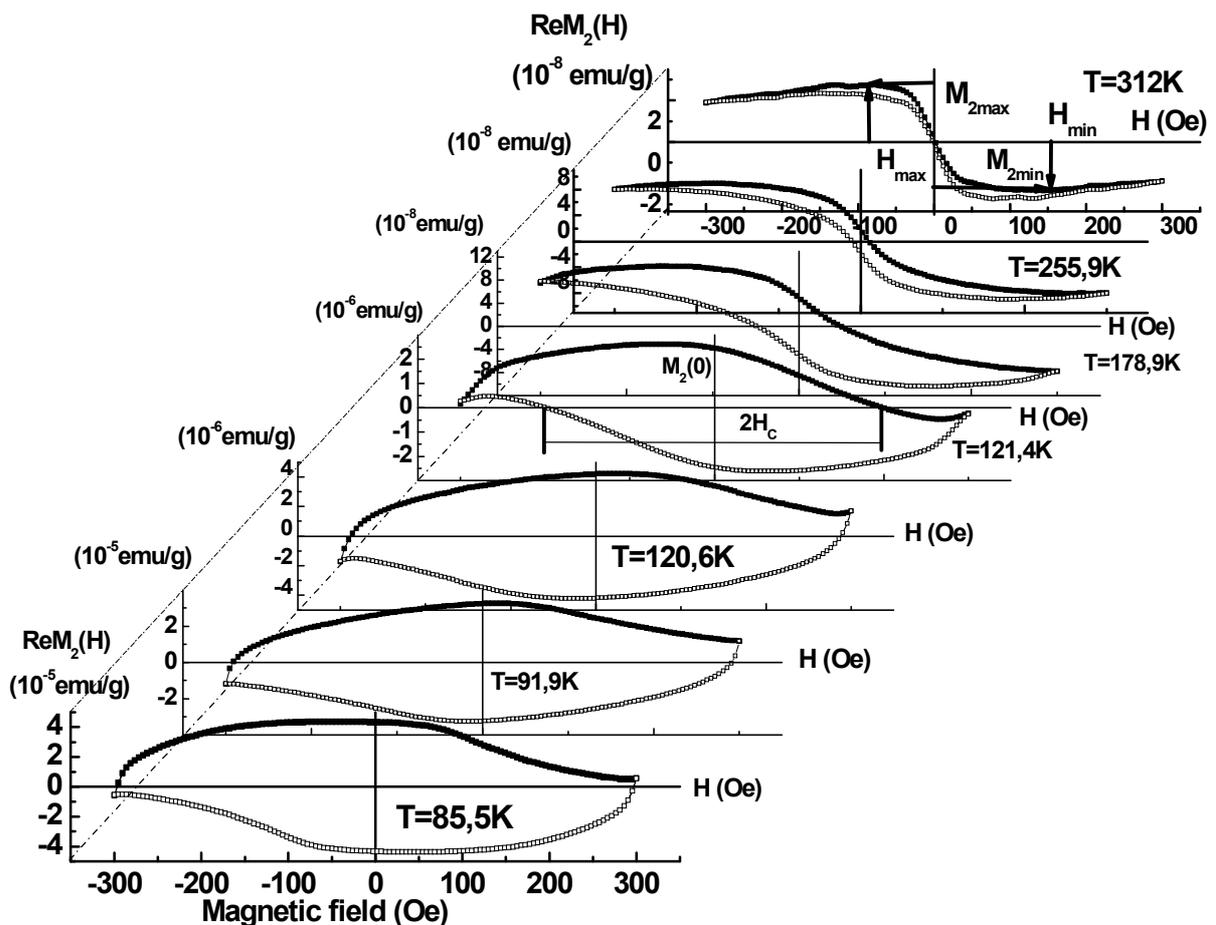

Fig.2. Real part of the second harmonic of magnetization as a function of the steady magnetic field at some temperatures. Direct (upper curves, closed squares) and reverse (down curves, open squares) H-scans are presented.

The M_2-measurements are performed in a temperature range 85 K ÷ 312 K. We present only ReM_2 data because a ratio ReM_2/ImM_2 ~ 10. Fig.2 shows some typical ReM_2 signals. A field hysteresis is observed in a finite range of H, $H_+ > |H| > H_-$ while ReM_2(0) = ReM_2(H) at $H = 0$ is equal zero (see Fig.2 at 312 K). This behavior persists down to 256 K where a small ReM_2(0) ~ 10^{-8} emu/g appears. The hysteresis loops are characterised by (i) the extremum values of ReM_2(H), M_{2max}, (ii) a position of these extremums H_{max}, and (iii) a coercivity H_C. Fig.1b presents these quantities and ReM_2(0) as a function of temperature. The M_2 response at H=0 provides clear evidence of a spontaneous magnetization of a sample [1]. The ReM_2(0) increases weakly with decreasing temperature down to 130 K, and then grows sharply (about two orders of magnitude) at a small reducing of temperature from 130 K to 117 K. The latter effect is associated with development of the F ordering, and suggests, at least, the weak first order transition at $T_C \approx$ 130 K, since the ReM_2(0) reveals the same order of the jumps at the appearance of F ordering in our sample and the $x = 0.45$ compound that exhibits the strong first order transition to the uniform F state [13]. In its turn, the small ReM_2(0) signal above 130 K can be related to the WF which appears at development of AF-ordering below $T_N{}^* \approx$ 256 K. The similar behavior of ReM_2(0) was also observed in the $x = 0.4$ [1] and $x = 0.45$ [13] compounds. The AF ordering is expected to be A-type since the M_2 signal is close to that found for the $x = 0.4$ and $x = 0.45$ manganites in the O' structure. For instance, the signals have the same order of ReM_2(0), ReM_{2max} and H_{max} at 160 K. The AF ordering above T_N is not registered by the neutron diffraction in the $x = 0.4$, 0.45 and our sample probably due to a small amount of this phase.

A discontinuity in H_{max}(T) and H_C(T) behavior at T=120K is associated with transformation of hysteresis loop (see Fig.2) due to development of the Jahn-Teller distortions (see Ref.3). Note that H-hysteresis of ReM_2(H) in the finite region of H, which is observed above 256 K, suggests the first order transition. This work was supported by joint Russian-Belorussian Foundation for Basic Research, Grants No 04-02-81051 RFBR-Bel2004_a and F04R-076.

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